Highly flexible and efficient top-emitting organic light-emitting devices with ultrasmooth Ag anode

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We demonstrate highly flexible and efficient top-emitting organic light-emitting devices (TOLEDs) by using an ultrasmooth Ag anode. A template-stripping process has been employed to create the ultrasmooth Ag anode on a photopolymer substrate. The flexible TOLEDs obtained by this method keep good electroluminescence properties under a small bending radius and after repeated bending. The efficiency of the flexible TOLEDs is improved by 60% compared with the conventional TOLEDs deposited on Si substrate due to the enhanced hole injection from the ultrasmooth anode. © 2012 Optical Society of America

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Organic light-emitting devices (OLEDs), particularly flexible OLEDs, have been attracting increasing attention owing to their advantages of being thin, lightweight, and ultraportable [1,2]. Flexible substrates are required for the realization of the flexible OLEDs. Among the substrates used for the flexible OLEDs, ultrathin glass sheet [3] has limited flexibility and is very brittle, and metal foil [4] is hard to handle in multiple bending. As a result, plastic film [1,2,5], e.g., polyethylene terephthalate, has become the most commonly used flexible substrate. Another key component in flexible OLEDs is the flexible electrodes. Indium-tin-oxide (ITO) is the traditionally used transparent anode material. Unfortunately, ITO has a high cost due to the scarcity of indium. Diffusion of indium into the adjacent organic films is another problem, which has been found to be correlated with the decay of the device performance [6]. ITO is especially not an ideal choice for flexible OLEDs because of its poor mechanical robustness [7–9] and manufacture incompatible with the plastic substrate due to the high-temperature deposition process [10]. Metal thin films have been adopted as one of the candidates to replace ITO [11,12], due to their high transparency and conductivity and their simple deposition by thermal evaporation. Moreover, top-emitting OLEDs (TOLEDs) can be easily realized by using the metal films as highly reflective bottom electrodes. However, the inherently rough surface of the metal films deposited by evaporation due to polycrystalinity is an obstacle to high performance of the OLEDs, because smoothness of the metal/organic interface is a key issue for both efficient carrier injection and long-term stability of the OLEDs [13,14].

Template stripping has been demonstrated to be a simple and effective technique to generate smooth metallic films [15–17]. Typically, solids with ultrasmooth surfaces, such as mica, glass, and silicon, are used as master templates. With subsequent metal deposition on the templates, a smooth surface at the metal/template interface is formed, which can be peeled off using a backing layer. Although the evaporated metal film has a rough surface after deposition, the smoothness of the opposite interface is near that of the templates. This method exhibits particular advantage in flexible OLEDs because not only can the metal smoothness be improved, but the backing layer itself is flexible and can be used as the substrate. However, further application of this method in flexible OLEDs has not yet been explored.

In this Letter, silver (Ag) film with ultrasmooth surface morphology has been fabricated on plastic substrate by employing the template stripping technique and has been used as the anode in flexible TOLEDs. The TOLEDs have shown superiority in both flexibility and mechanical robustness. Moreover, the efficiency is 60% enhanced compared with a conventional TOLED due to the enhanced hole injection at the ultrasmooth Ag/organic interface.

The fabrication process of ultrasmooth Ag anode on plastic substrate is shown in Fig. 1. At first, a cleaned silicon (Si) template was loaded into a thermal evaporation chamber. An 80 nm thick Ag was grown at a rate of 1 Å/s

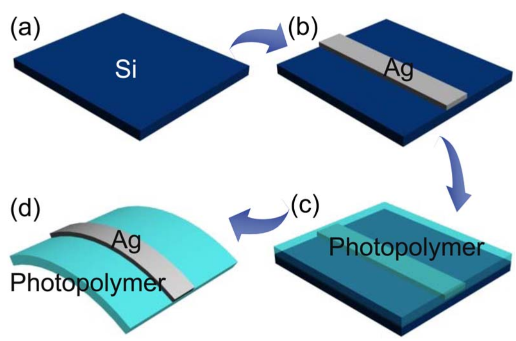

Fig. 1. (Color online) Scheme of fabricating ultrasmooth Ag film by template stripping. (a) A cleaned Si substrate is prepared as the template. (b) Ag film is deposited onto the Si template by thermal evaporation. (c) Photopolymer is spin-coated as a backing film. (d) The Ag film adhered to the cured photopolymer film is stripped.

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at a base pressure of 5×10^{-4} Pa. Then, a 1 mm thick photopolymer (NOA63, Norland) was spin coated onto the template deposited with the Ag film for 20 s at 1000 rpm and exposed to an ultraviolet light source for 8 min. The power of the light source is 100 W. At last, the cured photopolymer film can be peeled off, due to the better adhesion between Ag and the cured photopolymer compared with the Ag and Si substrate, so that the flexible substrate with ultrasmooth Ag was obtained. The repeatability of this process is fine over large areas.

The template-stripped Ag surface has a roughness that can approach that of the Si template, as confirmed by atomic force microscopy (AFM; iCON, Veeko). The AFM images of the surface morphology for Si and both sides of Ag film are shown in Fig. 2. The root mean square (rms) roughnesses for the Si template, the template-stripped, and as-evaporated Ag surfaces are 0.44, 0.625, and 1.36 nm, respectively. The roughness of the template-stripped Ag film is comparable to that of the template and is much improved compared with that of the as-evaporated side.

The utilization of the ultrasmooth Ag film as anode of the OLEDs has a benefit to form an efficient hole-injection contact between the anode and the hole-transport layer, so that the hole injection can be enhanced. The hole-only devices with both the as-deposited Ag anode on the conventional Si substrate and the ultrasmooth template-stripped Ag anode on the peeled-off photopolymer substrate were fabricated to examine the role of ultrasmooth Ag film in the hole-injection ability of the TOLEDs. The stack of structure is Ag(80 nm)/MoO_3 (4 nm)/4, 4', 4"-tris(3-methylphenylphe-nylamino) triphenylamine (m-MTDATA) (30 nm)/N, N'-diphenyl-N, N"-bis (1, 1'-biphenyl)-4, 4'-diamine (NPB) (70 nm)/Ag (20 nm). MoO_3 is a commonly used anodic buffer in TOLEDs [18]. m-MTDATA and NPB are hole-injection and hole-transport layers, respectively. The electron injection from the Ag cathode to the NPB is prohibited, because the work function of Ag is around 4.5 eV, while the lowest occupied molecular orbital level of the NPB is around 2.4 eV, and there exists a very large injection barrier. Here, all layers were deposited by thermal evaporation in a high vacuum system at a rate of 1 Å s^{-1} at a base pressure of 5×10^{-4} Pa.

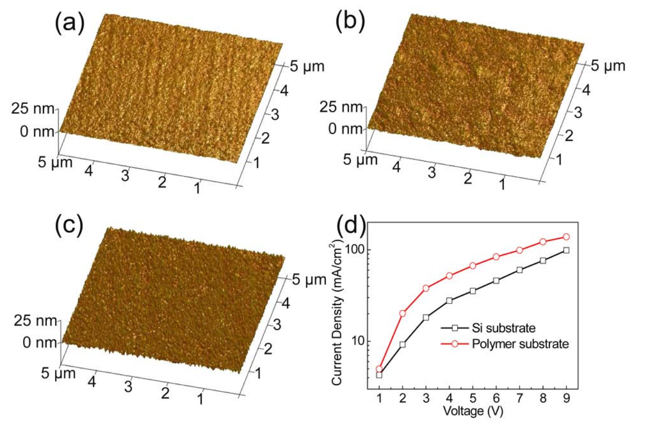

Fig. 2. (Color online) AFM images of (a) Si surface, (b) template-stripped Ag surface, and (c) as-evaporated Ag surface. (d) Current density-voltage characteristics of hole-only devices on Si and stripped photopolymer substrates.

The active area of the device is 2×2 mm^2. The current density (J)-voltage (V) characteristics of the devices were measured by a Keithley 2400 programmable voltage-current source. All of the measurements were conducted in air at room temperature. Figure 2(d) shows the J-V curves of the hole-only devices. It can be seen that the current density of devices with the ultrasmooth Ag anode is obviously higher than that of the devices with the as-deposited Ag anode. It confirms that an ultrasmooth Ag anode is crucial in enhancing the hole injection of the TOLEDs. Moreover, the ultrasmooth Ag anode has the additional benefit of reducing short circuits between the two electrodes that sandwich the thin organic stacks, so that improved device stability can be expected.

To demonstrate the effect of the ultrasmooth Ag anode on the electroluminescent (EL) performance of the TOLEDs, the TOLEDs with the template-stripped Ag anode on the peeled-off substrate and the as-deposited anode on the Si substrate were fabricated under identical process conditions. Tris-(8-hydroxyquinoline) aluminum (Alq_3) was used as an emitting layer. The device structure is Ag(80 nm)/MoO_3 (4 nm)/m-MTDATA (30 nm)/NPB (20 nm)/Alq_3 (50 nm)/LiF (1 nm)/Al (1 nm)/Ag (20 nm). The EL performance of the devices was measured by a Keithley 2400 programmable voltage-current source and Photo Research PR-655 spectrophotometer. The EL performance of TOLEDs are compared and shown in Fig. 3. It can be seen that TOLEDs fabricated on the peeled-off photopolymer substrate exhibit obvious improvement. Its maximum current efficiency is 10.5 cd/A, while it is 6.6 cd/A on the conventional Si substrate, which corresponds to a 60% enhancement. These improvements obviously originate from hole-injection enhancement as a result of lowered surface roughness of the Ag anode.

A bending test has been conducted to evaluate the flexibility of the TOLEDs. The devices operating at 6 V with different bending radii are shown in Figs. 4(a)–4(g). The bending radius is decreased by folding the substrate in a U shape and decreasing the distance between the two substrate edges. The operating device is free of cracks and dark spots even under an almost completely folded bending [Fig. 4(g)]. The mechanical robustness of the flexible TOLEDs is further investigated by measuring its EL performance after repeated bending. As shown in Fig. 4(h), no obvious deterioration can be observed in the luminance and efficiency after repeated bending. Moreover, the EL spectra and J-V curves are almost identical over up to 100 bending cycles, as shown in the inset of Fig. 4(h). The above results demonstrate that the

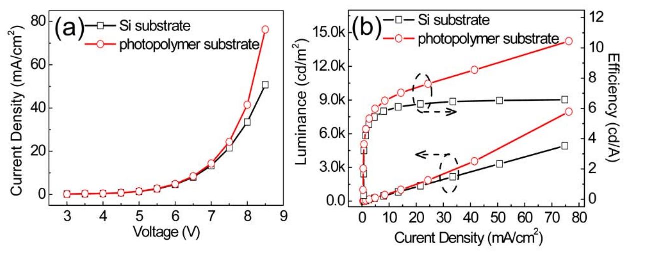

Fig. 3. (Color online) EL performance of TOLEDs on Si and peeled-off photopolymer substrates. (a) Current density-voltage and (b) luminance-current density-efficiency characteristics.

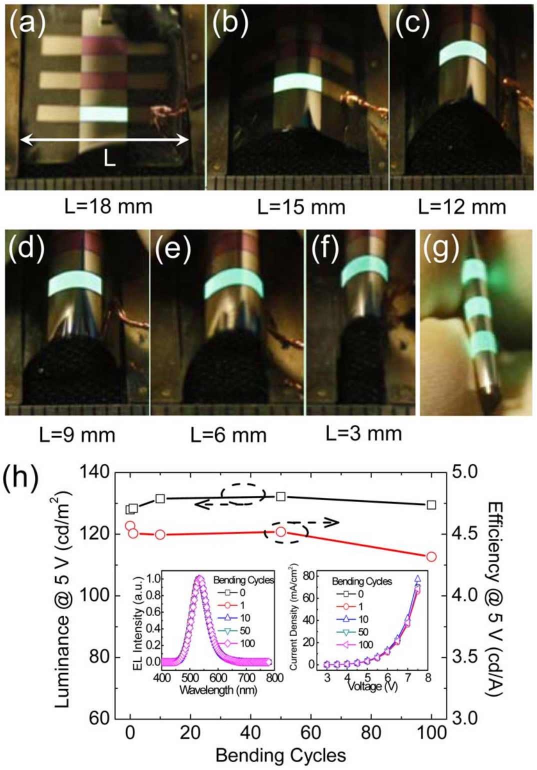

Fig. 4. (Color online) Flexibility and mechanical robustness of the flexible TOLEDs. (a)–(g) Photographs of the flexible TOLEDs at different bending radius. (h) Luminance and efficiency at 5 V as a function of the number of bending cycles. Inset in (h): Comparison of EL spectra and J-V characteristics before and after repeated bending.

TOLEDs are not only highly flexible but highly mechanically robust.

In summary, a template-stripping technique has been employed to create an ultrasmooth Ag anode on a flexible substrate for realizing highly flexible and efficient TOLEDs. The TOLEDs exhibit enhanced EL performance due to the ultrasmooth anode-induced improved hole injection. The high mechanical robustness of the flexibility has been demonstrated by conducting the bending test. From this study, we have confirmed that template stripping is an efficient method to realize flexible TOLEDs with high performance, which are promising candidates for flexible display and lighting.

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